

NASA TT F-10, 466

INVESTIGATION OF RECRYSTALLIZATION OF THIN FILMS  
UNDER THE EFFECT OF ELECTRON BOMBARDMENTA. N. Pilyankevich, V. P. Zakharov,  
and  
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Translation of "Issledovaniye rekristalizatsii tonkikh  
plenok pod vliyaniyem elektronnoy bombardirovki" Izvestia  
AN SSSR, Seriya Fizicheskaya, vol. 30, No. 5, p. 789-792,  
May 1966.

FACILITY FORM 602

N68-17613	
(ACCESSION NUMBER)	(THRU)
5	1
(PAGES)	(CODE)
✓	26
(NASA CR OR TMX OR AD NUMBER)	(CATEGORY)

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION  
WASHINGTON  
NOVEMBER 1966

INVESTIGATION OF RECRYSTALLIZATION OF THIN FILMS  
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Comparative study of the change in structure of thin films of germanium and silicon exposed to electron bombardment and thermal annealing. It is found that the temperature gradient arising during electron irradiation and the motion of the temperature front due to the thermal conductivity of the film results in recrystallization and the formation of large single crystals at thicknesses an order less than those observed by Gilbert et al. It is shown that, in contrast to previously prevailing opinion, recrystallization can occur in covalent crystals in the solid phase.

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The paper describes investigations of the structural changes of germanium and silicon films of a thickness on the order of 500 Å, obtained by spraying in vacuum at a pressure of  $1 \times 10^{-4}$  mm Hg, under electron bombardment and for comparison, under the effect of thermal annealing performed directly in an electron microscope.

Irradiation was performed by the following procedure: The condenser of the electron microscope was defocused, the limiting diaphragm was removed from it, then the condenser was rapidly refocused to concentrate the entire electron beam generated by the gun ( $U = 50$  kv,  $I = 25 \mu$  amp) onto a small area of the film, producing recrystallization in the latter. The process was practically instantaneous. It should be noted that under normal conditions no changes in a sample were observed for exposures lasting up to 30 min.

The initial sprayed film (both germanium and silicon) is in a metastable quasiamorphous state -- its granular structure does not appear in an electron microscope; electron diffraction patterns reveal four strongly blurred halos to which the following interplanar spacings may be ascribed: 3.3, 1.8, 1.15, and 0.93 Å for germanium, and 3.14, 1.74, 1.11, and 0.92 Å for silicon. After irradiation, the structure of the film changes.

For short exposures to a concentrated electron beam, in the film (within the limits of one element of the supporting mesh) arise three clearly defined regions (Fig. 1): (1) a central region with fine equiaxial grains, (2) an intermediate region with acicular and dendritic single crystals distributed

\*Numbers given in the margin indicate the pagination in the original foreign text.

along the radius from the periphery to the center of the mesh and measuring up to  $10^4$  ; at the edge of this region, the single crystals are also oriented but are distributed along its perimeter, and (3) a peripheral region in which the initial film structure remains unchanged. The electron diffraction pattern of such a recrystallized film has the form of a clearly defined diffraction spectrum with more than 40 lines.

The proof that the acicular formations are single crystals was obtained by the dark-field and microdiffraction methods. Figure 2 shows a photomicrograph obtained by the dark-field method, in which an area with equal orientation of the crystallographic planes was illuminated, and a low-energy electron diffraction pattern obtained by microdiffraction from an acicular single crystal (axis of the crystal zone [101] coincides with the direction of the electron beam).

This type of change in the crystalline structure of quasi-amorphous films can be explained in terms of the appearance of a radial temperature gradient caused by good heat transfer over the mesh to the massive film holder and by the motion of the temperature front owing to the thermal conductivity of the film. This explains the peculiar "zone-type" mechanism of the growth of germanium and silicon single crystals in the solid phase.

Subsequent long-term exposure of the irradiated film to the beam leads to heating of both the film holder and the mesh, an increase in temperature of the peripheral region, a decrease in the temperature gradient, and to the formation of fine equiaxial crystals analogous to those that form in the central region of the mesh element, where the temperature created at the initial moment of irradiation exceed the recrystallization temperatures of germanium and silicon (Fig. 1,b).

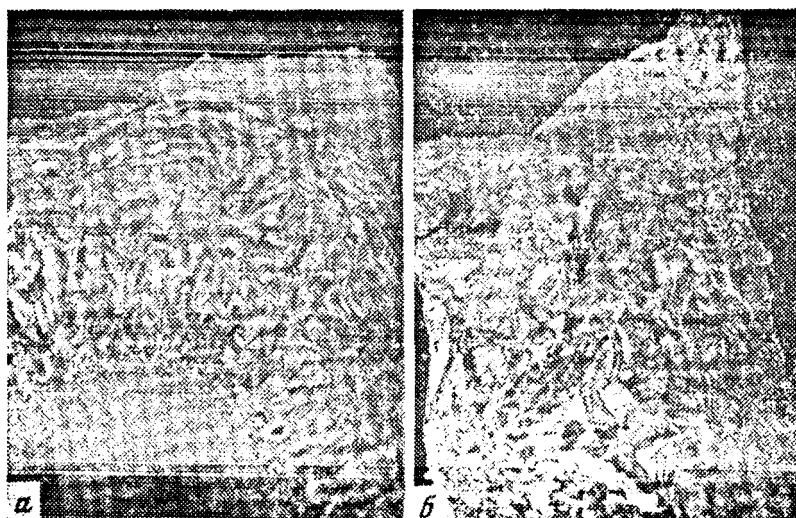


Fig. 1. Electron photomicrographs of a germanium film.  $1 \times 1400$ .  
a) short-term irradiation, b) prolonged irradiation

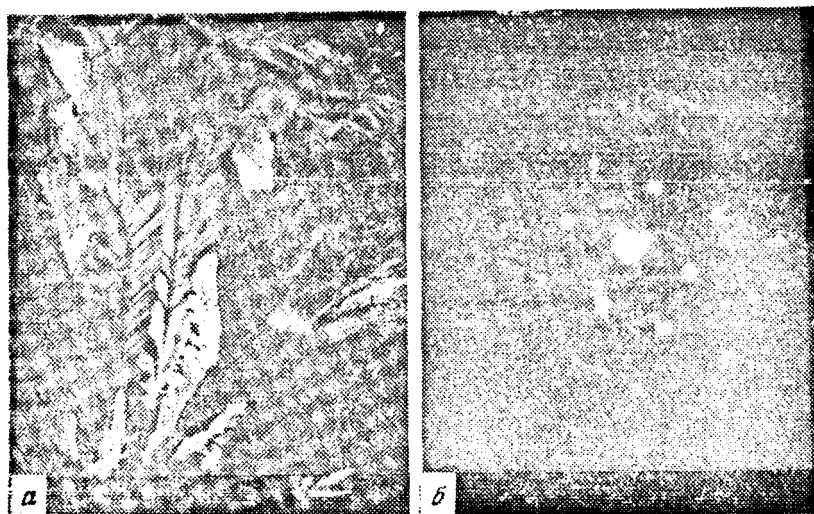


Fig. 2. a) Dark-field image of the recrystallized area of a germanium film.  $1 \times 5200$ . b) low-energy electron diffraction pattern from an acicular single crystal

In the case where the sprayed films are subjected to thermal heating directly in an electron microscope, the increase in film temperature is caused<sup>/791</sup> primarily by the thermal conductivity of the film; no appreciable temperature gradients arise in the process, which recrystallization leads to the formation of initially fine equiaxial crystals, the size of which increases later due to collective recrystallization (Fig. 3).

Publications are known (ref. 1), in which a coarse-grain oriented structure of thin films of some metals (Ge, Bi, In) sprayed onto a glass substrate was obtained by electron bombardment, using a movable electron beam to produce a temperature gradient. As a result of solid-phase recrystallization, large single crystals formed in the films. It was noticed that this process occurred only at film thicknesses in excess of  $10,000 \text{ \AA}$ .

In our experiments, the temperature gradient created by electron bombardment<sup>/792</sup> and by the motion of the temperature front owing to the thermal conductivity of the film lead to recrystallization accompanied by the formation of large single crystals at film thicknesses smaller by an order of magnitude than the just mentioned ones.

Another interesting feature of the result obtained is that, contrary to existing opinion according to which recrystallization in the solid phase cannot take place in covalent crystals (ref. 2), in our tests, this process does take

place, as can be clearly seen from electron-microscope observations of the films during the irradiation process.

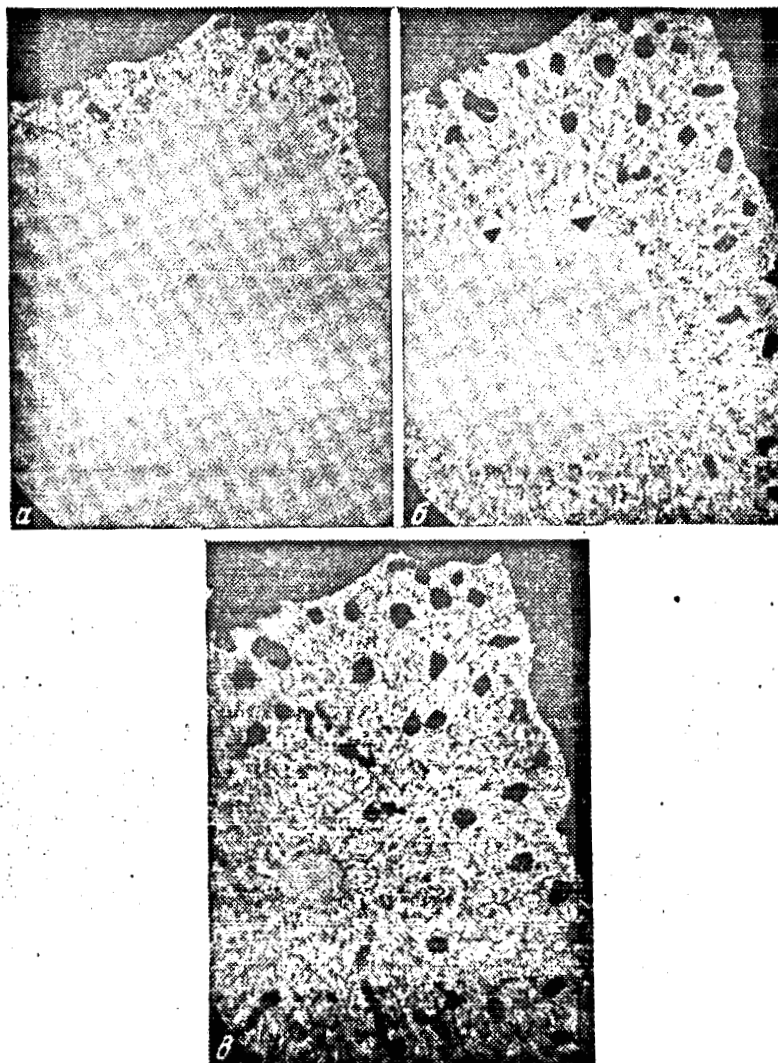


Fig. 3. Recrystallization phases in the case of thermal annealing of a sprayed germanium film.  $1 \times 100$ . Annealing temperature -  $300^{\circ}\text{C}$ . Annealing time: a) 18 min, b) 25 min, c) 35 min.

In conclusion we would like to note an interesting peculiarity of the electron diffraction patterns of the recrystallized films. In addition to the diffraction lines characteristic of the diamond-type germanium and silicon lattice, the electron diffraction patterns reveal many lines pertaining to an fcc lattice, which are forbidden for a diamond-type lattice by the structure factor. These are the lines with the indices (200), (222), (420), (600), (622), and so on. Usually, germanium electron diffraction patterns reveal only one forbidden line -- the (222) line (the strongest line in our tests) (ref. 3). Apparently, this effect can be explained in terms of the phenomenon of multiple diffraction

in which case two allowed images, combining vectorially in an inverse space, may produce a normally forbidden resulting image -- for example:  
 $[111] + [111] \rightarrow [200]$ ;  $[311] + [111] \rightarrow [200]$ ;  $[331] + [111] \rightarrow [420]$ , and so forth.

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Translated for National Aeronautics and Space Administration by  
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